

original claim 2 has not been reintroduced as a new claim)

Please cancel claims 30-45 and 49-50 without prejudice and disclaimer.

Please add the following new claims:

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B/ 51. (NEW) A method for MRI imaging comprising administering to a patient an MRI contrast agent, comprising a perfluoroalkyl-containing metal complex that has a critical micelle formation concentration  $< 10^{-3}$  mol/l, a hydrodynamic micelle diameter ( $2 R_h$ )  $> 1$  nm and a proton relaxivity in plasma ( $R^1$ )  $> 10$  l/mmol's and conducting MRI imaging whereby plaque, or necrotic tissue are visualized or necroses and tumors are independently visualized.

52. (NEW) A method according to claim 51, wherein necrotic tissue is visualized.

53. (NEW) A method according to claim 51, wherein necroses or tumors are independently visualized.

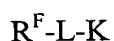
54. (NEW) A method according to claim 51, wherein the metal complex has a micelle formation concentration of  $< 10^{-4}$  mol/l.

55. (NEW) A method according to claim 51, wherein the metal complex has a hydrodynamic micelle diameter of  $> 3$  nm.

56. (NEW) A method according to claim 51, wherein the metal complex has a proton relaxivity in plasma of  $> 13$  l/mmol's.

57. (NEW) A method according to claim 51, wherein the perfluoroalkyl-

containing metal complex is a compound of formula I



I

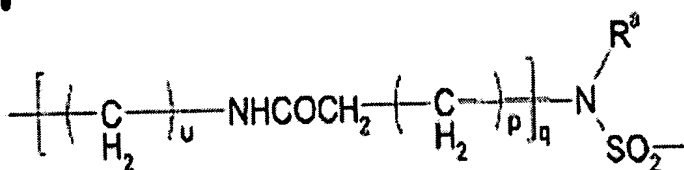
in which

$R^F$  is a perfluorinated, straight-chain or branched carbon chain with formula - $C_nF_{2n}E$ , in which

$E$  is a terminal fluorine, chlorine, bromine, iodine or hydrogen atom and  $n$  is a number from 4-30,

$L$  is a direct bond, a methylene group, an -NHCO- group, a group

Bl  
Cmt



whereby  $p$  is a number from 0 to 10, and  $q$  and  $n$ , independently of one another, are 0 or 1, and  $R^a$  is a hydrogen atom, a methyl group, a -CH<sub>2</sub>-OH group, a -CH<sub>2</sub>-CO<sub>2</sub>H group or a C<sub>2</sub>-C<sub>15</sub> alkyl, which optionally is interrupted by 1 to 3 oxygen atoms, 1 to 2 CO groups or an optionally substituted aryl group and/or is substituted with 1 to 4 hydroxyl groups, 1 to 2 C<sub>1</sub>-C<sub>4</sub> alkoxy groups, 1 to 2 carboxy groups, or a group -SO<sub>3</sub>H,

or

$L$  is a straight-chain, branched, saturated or unsaturated C<sub>2</sub>-C<sub>30</sub> carbon chain, which optionally contains 1 to 10 oxygen atoms, 1 to 3 -NR<sup>a</sup> groups, 1 to 2 sulfur atoms, a piperazine group, a -CONR<sup>a</sup> group, an -NR<sup>a</sup>CO group, an -SO<sub>2</sub> group, an -NR<sup>a</sup>-CO<sub>2</sub> group, 1 to 2 CO groups, a group -CO-N-T-N(R<sup>a</sup>)-SO<sub>2</sub>-R<sup>F</sup>, or 1 to 2 optionally substituted aryls and/or is interrupted by these groups and/or is optionally

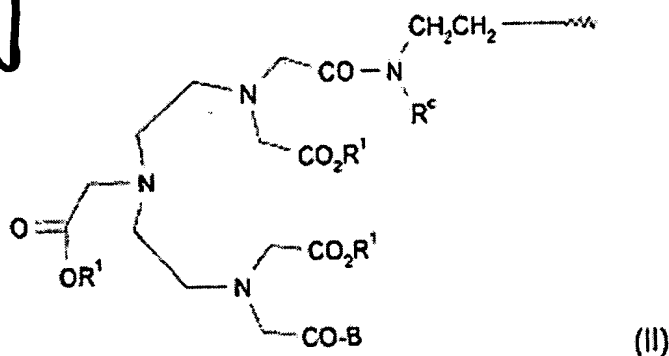
substituted with 1 to 3  $-OR^a$  groups, 1 to 2 oxo groups, 1 to 2  $-NH-COR^a$  groups, 1 to 2  $-CONHR^a$  groups, 1 to 2  $-(CH_2)_p-CO_2H$  groups, 1 to 2 groups  $-(CH_2)_p-(O)_q-CH_2CH_2-R^F$ ,

whereby

$R^a$ ,  $R^F$  and p and q have the above-indicated meanings, and

T is a  $C_2-C_{10}$  chain, which optionally is interrupted by 1 to 2 oxygen atoms or 1 to 2  $-NHCO$  groups,

K is a complexing agent or metal complex of formula II



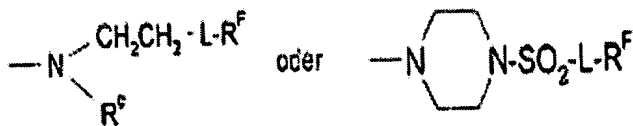
in which  $R^c$ ,  $R^1$  and B are independent of one another,

and

$R^c$  is  $R^a$  or is  $-(CH_2)_m-L-R^F$ , whereby m is 0, 1 or 2, and L and  $R^F$  have the above-mentioned meaning,

$R^1$ , independently of one another, is a hydrogen atom or a metal ion equivalent of atomic numbers 22-29, 42-46 or 58-70,

B is  $-OR^1$ ,



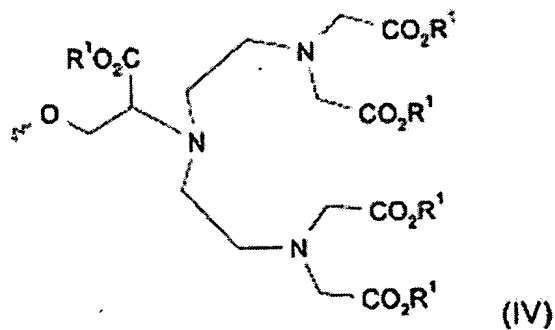
whereby  $R^1$ , L,  $R^F$  and  $R^c$  have the above-mentioned

K is a complexing agent or complex of formula III



or

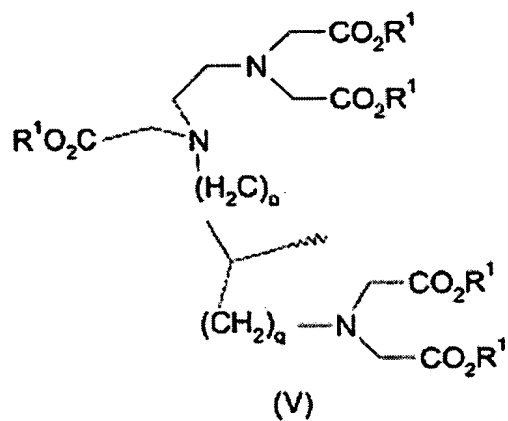
K is a complexing agent or complex of formula IV



in which  $R^1$  has the above-mentioned meaning

or

K is a complexing agent or complex of formula V

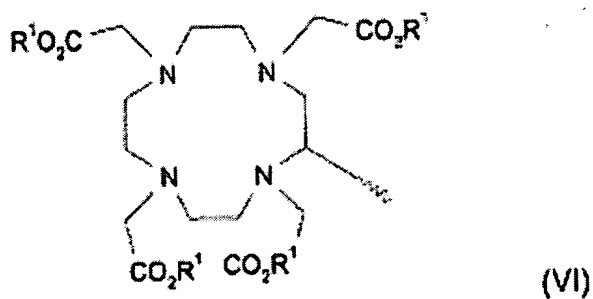


in which  $R^1$  has the above-mentioned meaning, and  $o$  and  $q$  stand for numbers

0 or 1, and yields the sum  $o + q = 1$ ,

or

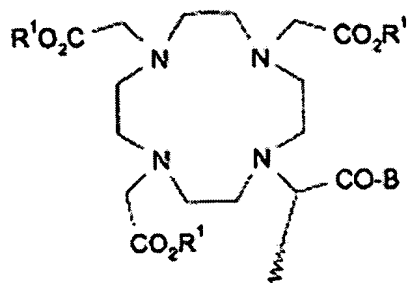
K is a complexing agent or complex of formula VI



in which  $R^1$  has the above-mentioned meaning

or

K is a complexing agent or complex of formula VII

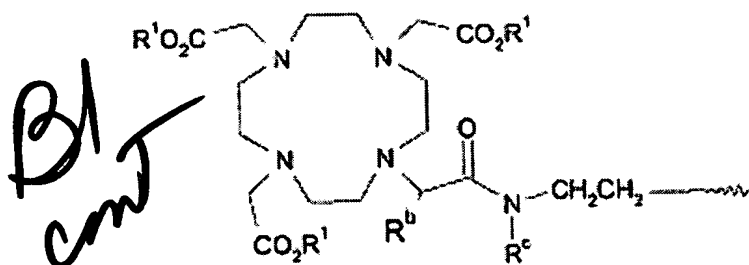


(VII)

in which R<sup>1</sup> and B have the above-mentioned meanings

or

K is a complexing agent or complex of formula VIII

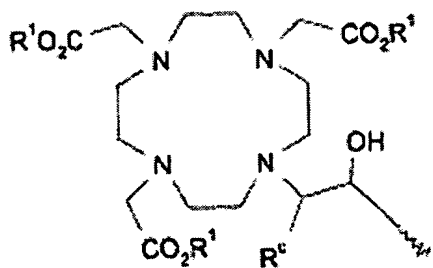


(VIII)

in which R<sup>c</sup> and R<sup>1</sup> have the above-mentioned meanings, and R<sup>b</sup> is R<sup>a</sup>

or

K is a complexing agent or complex of formula IX

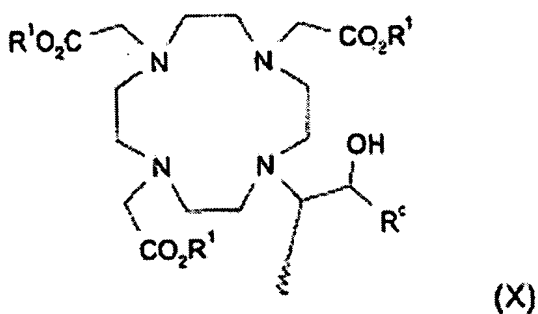


(IX)

in which R<sup>c</sup> and R<sup>1</sup> have the above-mentioned meanings,

or

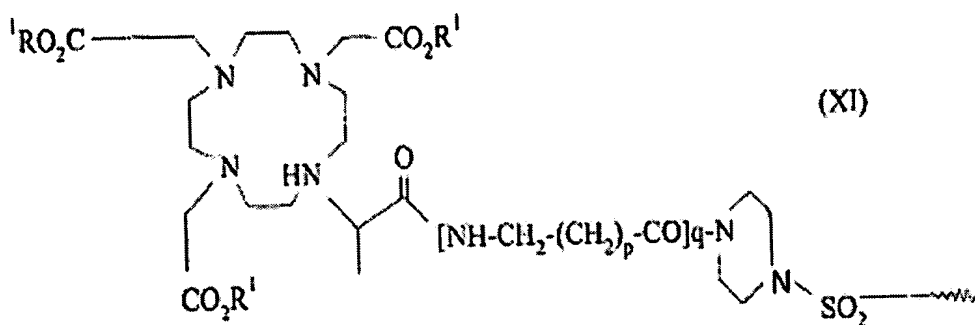
K is a complexing agent or complex of formula X



in which  $R^c$  and  $R^1$  have the above-mentioned meanings,

or

K is a complexing agent or complex of formula XI

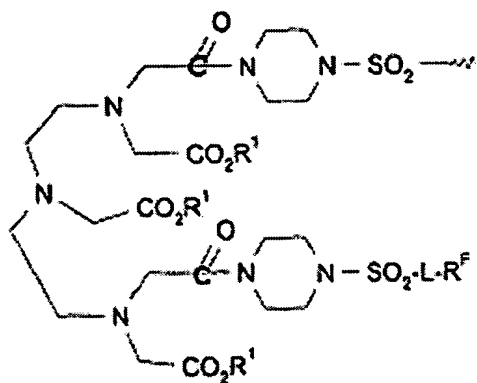


in which  $R^1$ ,  $p$  and  $q$  have the above-mentioned meanings,

and  $R^b$  has the meaning of  $R^a$ ,

or

K is a complexing agent or complex of formula XII

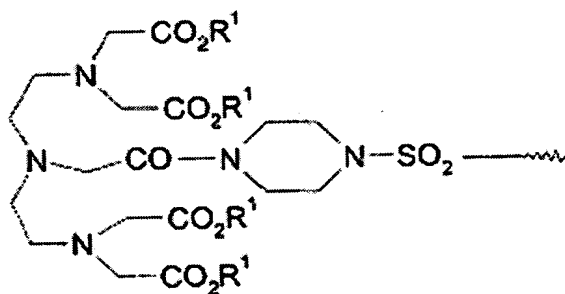


(XII)

in which L, R<sup>F</sup> and Z<sup>1</sup> have the above-mentioned meanings,

or

is a complexing agent or complex of formula XIII



(XIII)

in which R<sup>1</sup> has the above-mentioned meaning, or

K is a salt of one of the complexing agents or complexes of formula II to XIII with an organic and/or inorganic base or amino acid or amino acid amide.

58. (NEW) A method according to claim 57, wherein in the compound of formula I,

L is

$\alpha\text{-CH}_2\text{-}\beta$

$\alpha\text{-CH}_2\text{CH}_2\text{-}\beta$

$\alpha\text{-(CH}_2\text{)}_s\text{-}\beta$      $s = 3 - 15$

$\alpha\text{-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$

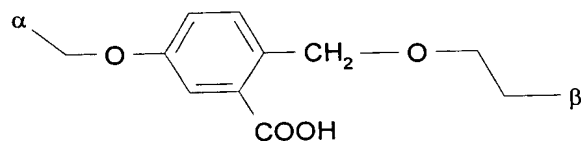
$\alpha\text{-CH}_2\text{-(O-CH}_2\text{-CH}_2\text{)}_t\text{-}\beta$      $t = 2 - 6$



$\alpha$ -CH<sub>2</sub>-NH-CO- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NH-CO-CH<sub>2</sub>-N(CH<sub>2</sub>COOH)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NH-CO-CH<sub>2</sub>-N(C<sub>2</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NH-CO-CH<sub>2</sub>-N(C<sub>10</sub>H<sub>21</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NH-CO-CH<sub>2</sub>-N(C<sub>6</sub>H<sub>13</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NH-CO-(CH<sub>2</sub>)<sub>10</sub>-N(C<sub>2</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NH-CO-CH<sub>2</sub>-N(-CH<sub>2</sub>-C<sub>6</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NH-CO-CH<sub>2</sub>-N(-CH<sub>2</sub>-CH<sub>2</sub>-OH)SO<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NHCO-(CH<sub>2</sub>)<sub>10</sub>-S-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>NHCOCH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>NHCO(CH<sub>2</sub>)<sub>10</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-C<sub>6</sub>H<sub>4</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-O-CH<sub>2</sub>-C(CH<sub>2</sub>-OCH<sub>2</sub>CH<sub>2</sub>-C<sub>6</sub>F<sub>13</sub>)<sub>2</sub>-CH<sub>2</sub>-OCH<sub>2</sub>-CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-NHCOCH<sub>2</sub>CH<sub>2</sub>CON-CH<sub>2</sub>CH<sub>2</sub>NHCOCH<sub>2</sub>N(C<sub>2</sub>H<sub>5</sub>)SO<sub>2</sub>C<sub>8</sub>F<sub>17</sub>  

$\downarrow$   
 $\text{CH}_2\text{-CH}_2\text{NHCOCH}_2\text{N(C}_2\text{H}_5\text{)-SO}_2\text{-}\beta$

 $\alpha$ -CH<sub>2</sub>-O-CH<sub>2</sub>-CH(OC<sub>10</sub>H<sub>21</sub>)-CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -(CH<sub>2</sub>NHCO)<sub>4</sub>-CH<sub>2</sub>O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -(CH<sub>2</sub>NHCO)<sub>3</sub>-CH<sub>2</sub>O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-OCH<sub>2</sub>C(CH<sub>2</sub>OH)<sub>2</sub>-CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$



$\alpha$ -CH<sub>2</sub>NHCOCH<sub>2</sub>N(C<sub>6</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -NHCO-CH<sub>2</sub>-CH<sub>2</sub>- $\beta$   
 $\alpha$ -NHCO-CH<sub>2</sub>-O-CH<sub>2</sub>CH<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>-N(CH<sub>2</sub>COOH)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>-N(C<sub>2</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>-N(C<sub>10</sub>H<sub>21</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>-N(C<sub>6</sub>H<sub>13</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-(CH<sub>2</sub>)<sub>10</sub>-N(C<sub>2</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>-N(-CH<sub>2</sub>-C<sub>6</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>-N(-CH<sub>2</sub>-CH<sub>2</sub>-OH)SO<sub>2</sub>- $\beta$   
 $\alpha$ -NH-CO-CH<sub>2</sub>- $\beta$

$\alpha$ -CH<sub>2</sub>-O-C<sub>6</sub>H<sub>4</sub>-O-CH<sub>2</sub>-CH<sub>2</sub>- $\beta$   
 $\alpha$ -CH<sub>2</sub>-C<sub>6</sub>H<sub>4</sub>-O-CH<sub>2</sub>-CH<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>2</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>6</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>10</sub>H<sub>21</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>6</sub>H<sub>13</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(C<sub>2</sub>H<sub>4</sub>OH)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(CH<sub>2</sub>COOH)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N(CH<sub>2</sub>C<sub>6</sub>H<sub>5</sub>)-SO<sub>2</sub>- $\beta$   
 $\alpha$ -N-[CH(CH<sub>2</sub>OH)<sub>2</sub>]-SO<sub>2</sub>- $\beta$  or  
 $\alpha$ -N-[CH(CH<sub>2</sub>OH)CH(CH<sub>2</sub>OH)]-SO<sub>2</sub>- $\beta$ ,

in which  $\alpha$  is the binding site to the complexing agent or metal complex K, and  $\beta$  is the binding site to the fluorine radical.

59. (NEW) A method according to claim 57, wherein the compound of formula I, is a compound in which n in formula -C<sub>n</sub>F<sub>2n</sub>E is a number from 4-15 and/or E is a fluorine atom.

60. (NEW) A method according to claims 57, wherein the compound of formula I is:

Gadolinium complex of 10-[1-methyl-2-oxo-3-aza-5-oxo-{4-perfluorooctylsulfonyl-piperazin-1-yl}-pentyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,

Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-oxa-10,10,11,11,12,12,13,13,14,14,15,15,16,16,17,17-heptadecafluoroheptadecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,

Gadolinium complex of 10-[2-hydroxy-4-aza-5,9-dioxo-9-{4-perfluorooctyl}-piperazin-1-yl]-nonyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,

Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-aza-7-(perfluorooctylsulfonyl)-nonyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,

Gadolinium complex of 10-[2-hydroxy-4-oxa-

1H,1H,2H,3H,3H,5H,5H,6H,6H-perfluorotetradecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,

Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-oxa-10,10,11,11,12,12,13,13,14,14,15,15,-16,16,17,17,18,18,19,19-henicosafuoro-nonadecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane,

Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-11-aza-11-(perfluorooctylsulfonyl)-tridecyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraazacyclododecane, or

Gadolinium complex of 10-[2-hydroxy-4-aza-5-oxo-7-aza-7-(perfluorooctylsulfonyl)-8-phenyl-octyl]-1,4,7-tris(carboxymethyl)-1,4,7,10-tetraaza-cyclododecane.

61. (NEW) A method according to claim 51, wherein the-perfluoroalkyl-containing metal complex, is a compound of formula Ia



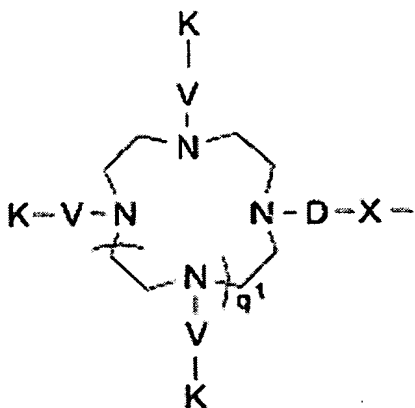
in which

A is a group that contains 2 to 6 metal complexes, which are bonded directly or via a linker to a nitrogen atom of an annular skeleton chain, and

$R^F$  is a perfluorinated, straight-chain or branched carbon chain with formula  $-C_nF_{2n}E$ , in which

E is a terminal fluorine, chlorine, bromine, iodine or hydrogen atom, and n is a number from 4-30,

whereby A has the following structure:



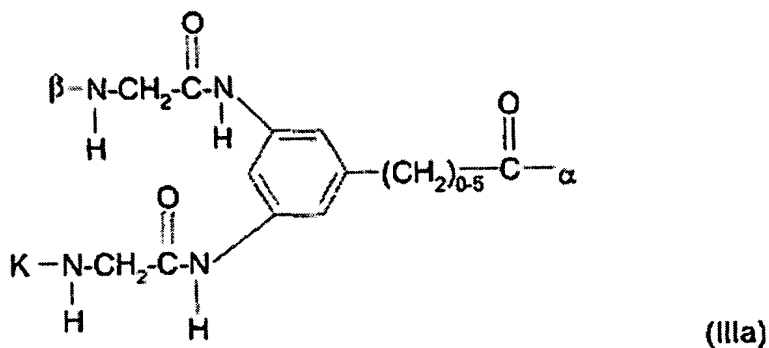
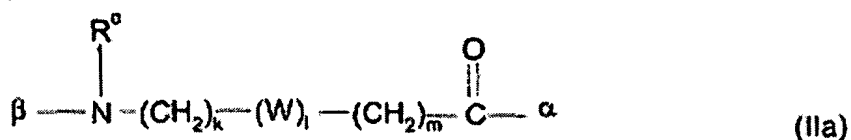
whereby

$q^1$  is 0, 1, 2 or 3,

K is a complexing agent or metal complex or a salts thereof with an organic and/or inorganic base or amino acid or amino acid amide,

X as the point of attachment to  $R^F$ , is a direct bond, a phenylene group or a  $C_1$ - $C_{10}$  alkylene chain, which optionally contains 1-15 oxygen atoms, 1-5 sulfur atoms, 1-10 carbonyl groups, 10-10 ( $NR^d$ ) groups, 1-2  $NR^dSO_2$  groups, 1-10  $CONR^d$  groups, 1 piperidine group, 1-3  $SO_2$  groups and/or 1-2 phenylene groups or optionally is substituted by 1-3 radicals  $R^F$ , in which  $R^d$  is a hydrogen atom, a phenyl group, benzyl group or a  $C_1$ - $C_{15}$  alkyl group, which optionally contains 1-2  $NHCO$ , 1-2  $CO$  groups, 1-5 oxygen atoms and optionally is substituted by 1-5 hydroxy, 1-5 methoxy, 1-3 carboxy, or 1-3  $R^F$  radicals,

V is a direct bond or a chain of formula IIa or IIIa:



in which

—  $\text{R}^e$  is a hydrogen atom, a phenyl group, a benzyl group or a  $\text{C}_1\text{-C}_7$  alkyl group, which optionally is substituted with a carboxy group, a methoxy group or a hydroxy group,

—  $\text{W}$  is a direct bond, a polyglycol ether group with up to 5 glycol units, or a group of formula IVa



in which  $\text{R}^h$  is a  $\text{C}_1\text{-C}_7$  carboxylic acid, a phenyl group, a benzyl group or a  $(\text{CH}_2)_{1-5}\text{-NH-K}$  group,

—  $\alpha$  is the binding to the nitrogen atom of the skeleton chain,  $\beta$  is the binding to complexing agent or metal complex  $\text{K}$ ,

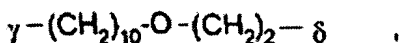
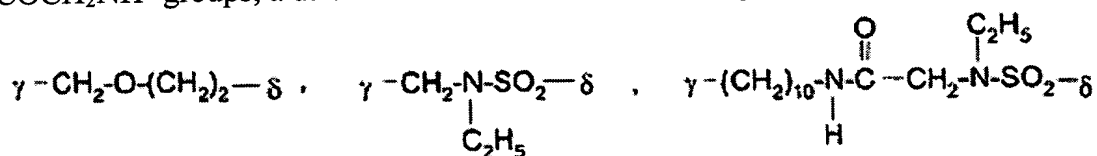
— and in which variables  $k$  and  $m$  stand for natural numbers between 0 and 10, and  $l$  is 0 or 1

and whereby

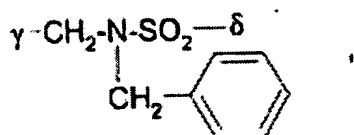
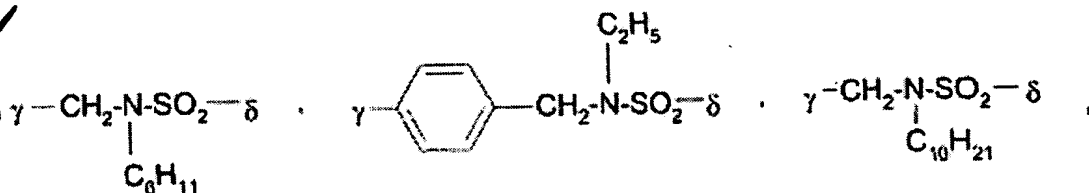
$\text{D}$  is a  $\text{CO}$  or  $\text{SO}_2$  group.

62. (NEW) A method according to claim 61, wherein the compound of formula Ia is a compound in which  $q^1$  is the number 1.

63. (NEW) A method according to claim 61, wherein the compound of formula Ia is a compound in which X is an alkylene chain, which contains 1-10 -CH<sub>2</sub>CH<sub>2</sub>O- groups or 1-5 -COCH<sub>2</sub>NH- groups, a direct bond or one of the following structures



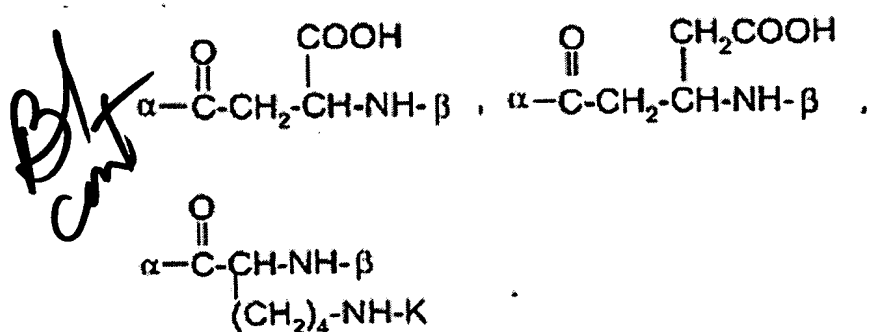
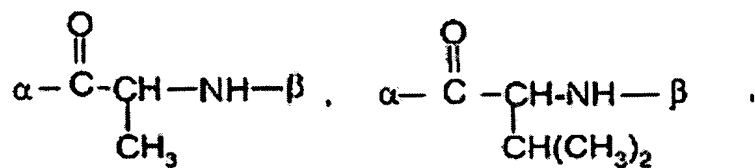
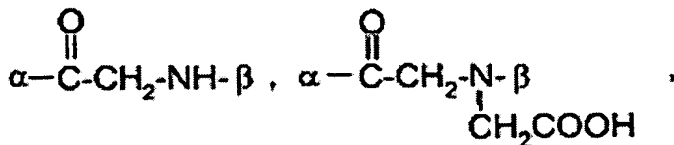
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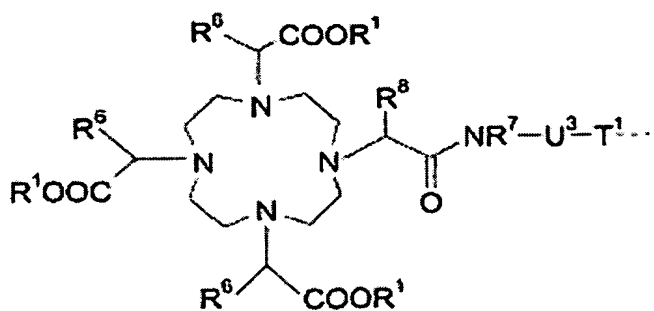
whereby

$\gamma$  binds to D, and  $\delta$  binds to R<sup>F</sup>.

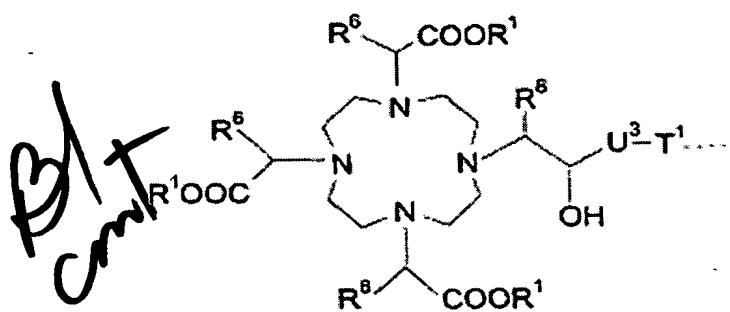
64. (NEW) A method according to claim 61, wherein the compound of formula Ia, is a compound in which V is a group with one of the following structures



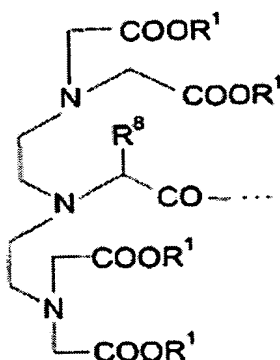
65. (NEW) A method according to claim 61, wherein the compound of formula Ia, is a compound in which K is a complexing agent or complex of formula Va, VIa, VIIa or VIIIa,



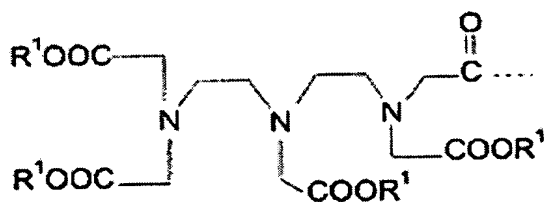
(Va)



(Via)



(VIIa)



(VIIIa)

whereby

$R^1$ , independently of one another, are a hydrogen atom or a metal ion equivalent of the elements of atomic numbers 23-29, 42-46 or 58-70,  
 $R^8$  is a hydrogen atom or a straight-chain, branched, saturated or unsaturated  $C_1$ - $C_{30}$  alkyl chain, which optionally is substituted by 1-5 hydroxy, 1-3



carboxy or 1 phenyl group(s) and/or optionally is interrupted by 1-10 oxygen atoms, 1 phenylene group or 1 phenylenoxy group,

$R^6$  are independently a hydrogen atom, a straight-chain or branched  $C_1-C_7$  alkyl radical, a phenyl radical or benzyl radical,

$R^7$  is a hydrogen atom, a methyl group or ethyl group, which optionally is substituted by a hydroxy group or carboxy group,

$U^3$  is a straight-chain, branched, saturated or unsaturated  $C_1-C_{20}$  alkylene group optionally containing 1-5 imino groups, 1-3 phenylene groups, 1-3 phenylenoxy groups, 1-3 phenylenimino groups, 1-5 amide groups, 1-2 hydrazide groups, 1-5 carbonyl groups, 1-5 ethylenoxy groups, 1 urea group, 1 thiourea group, 1-2 carboxyalkylimino groups, 1-2 ester groups, 1-1-0 oxygen atoms, 1-5 sulfur atoms and/or 1-5 nitrogen atoms, and/or optionally substituted by 1-5 hydroxy groups, 1-2 mercapto groups, 1-5 oxo groups, 1-5 thioxo groups, 1-3 carboxy groups, 1-5 carboxyalkyl groups, 1-5 ester groups and/or 1-3 amino groups, whereby the optionally contained phenylene groups can be substituted by 1-2 carboxy groups, 1-2 sulfone groups or 1-2 hydroxy groups

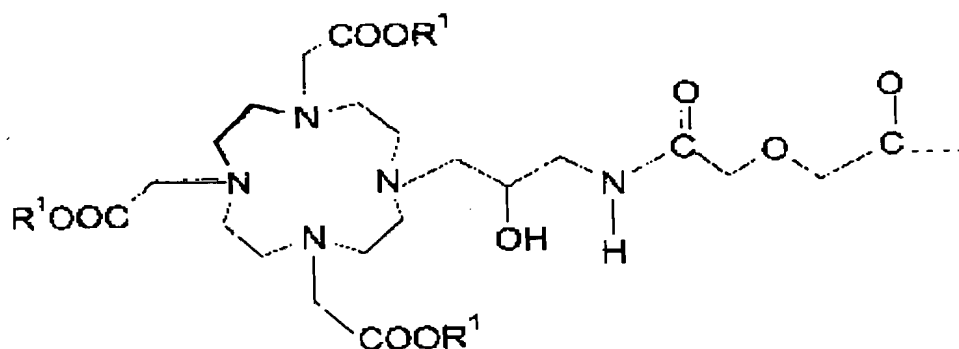
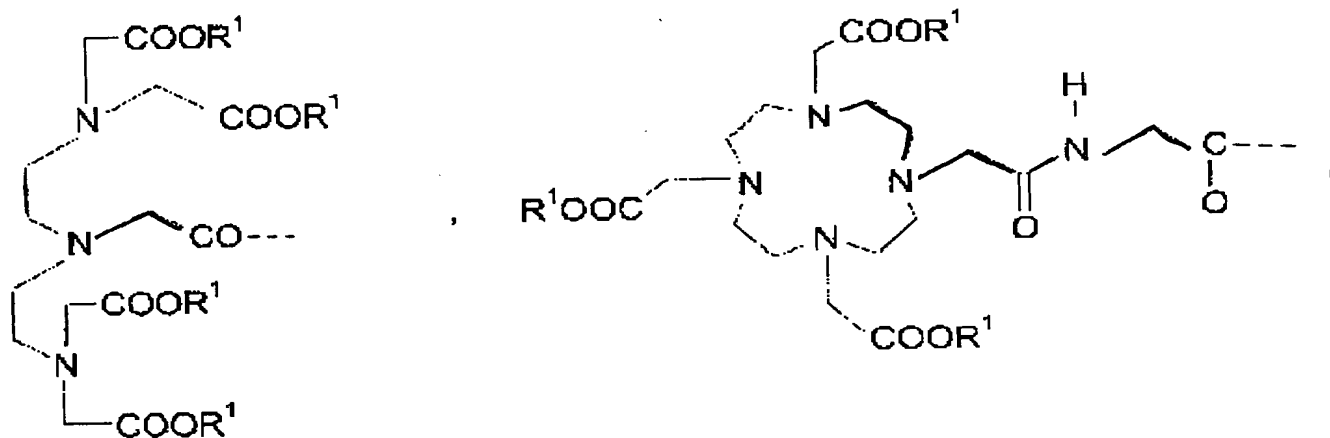
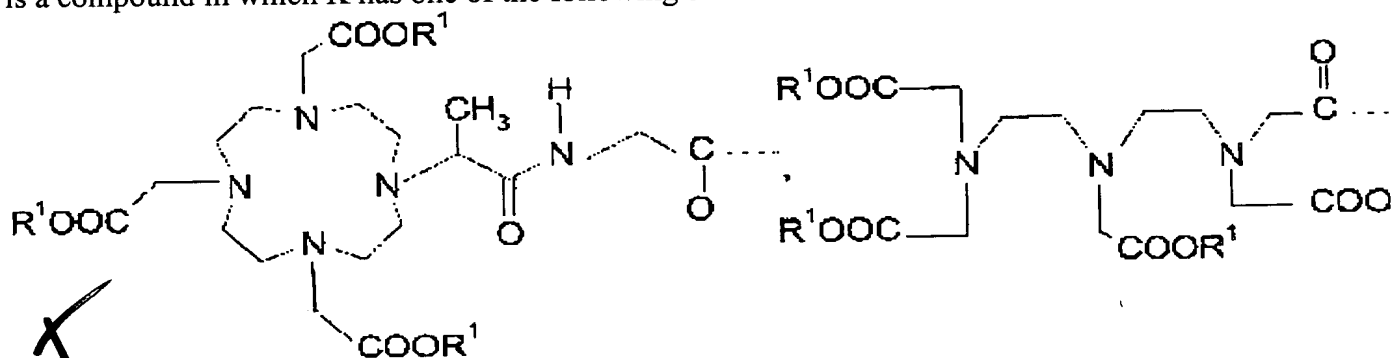
$T^1$  is a  $-CO-\beta$ ,  $-NHCO-\beta$  or  $-NHCS-\beta$  group, whereby  $\beta$  is the binding site to V.

66. (NEW) A method according to claim 65, wherein the  $C_1-C_{20}$  alkylene chain that is  $U^3$  contains the group  $-CH_2NHCO-$ ,  $-NHCOCH_2O-$ ,  $-NHCOCH_2OC_6H_4-$ ,  $-N(CH_2CO_2H)-$ ,  $-CH_2OCH_2-$ ,  $-NHCOCH_2C_6H_4-$ ,  $-NHCSNHC_6H_4-$ ,  $-CH_2OC_6H_4-$ , or  $-CH_2CH_2O-$  and/or is substituted by the group  $-COOH$  and/or  $-CH_2COOH$ .

67. (NEW) A method according to claim 65, wherein  $U^3$  is a  $-CH_2-$ ,  $-CH_2CH_2-$ ,  $-CH_2CH_2CH_2-$ ,  $-C_6H_4-$ ,  $-C_6H_{10}-$ ,  $-CH_2C_6H_4-$ ,  $-CH_2NHCOCH_2CH(CH_2CO_2H)-C_6H_4-$ ,  $-CH_2NHCOCH_2OCH_2-$ , or  $-CH_2NHCOCH_2C_6H_4-$

group.

68. (NEW) A method according to claim 61, wherein the compound of formula Ia, is a compound in which K has one of the following structures:



69. (NEW) A method according to claim 61, wherein the compound of formula Ia, is a compound in which the perfluoroalkyl chain is  $R^F$  is  $-C_6F_{13}$ ,  $-C_8F_{17}$ ,  $-C_{10}F_{21}$  or  $-C_{12}F_{25}$ .

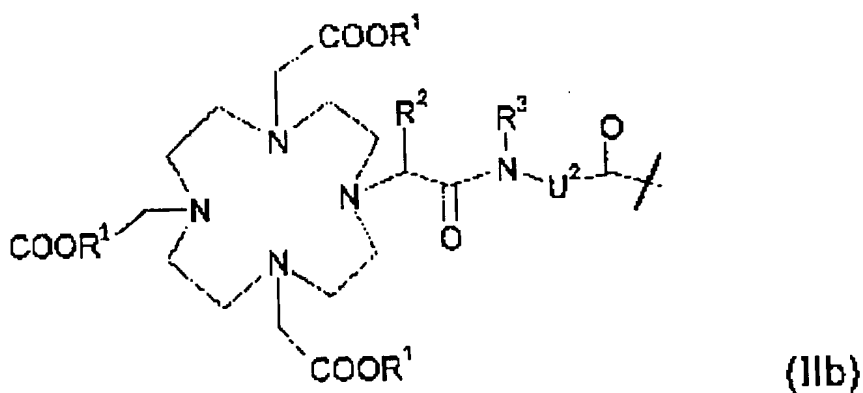
70. (NEW) A method according to claim 61, wherein the compound of formula Ia is 1,4,7-tris{1,4,7-tris(N-(carboxylatomethyl)-10-[N-1-methyl-3,6-diaza-2,5,8-trioxooctane-1,8-diyl])-1,4,7,10-tetraazacyclododecane, Gd complex}-10-[N-2H,2H,4H,4H,5H,5H-3-oxa-perfluorotridecanoyl]-1,4,7,10-tetraazacyclododecane.

71. (NEW) A method according to claim 51, wherein the perfluoroalkyl-containing metal complex, is a compound of formula Ib



in which

K is a complexing agent or a metal complex of formula IIb



whereby


$R^1$  is a hydrogen atom or a metal ion equivalent of atomic numbers 23-29, 42-46 or 58-70,

$R^2$  and  $R^3$  are independently a hydrogen atom, a  $C_1$ - $C_7$  alkyl group, a benzyl

group, a phenyl group,  $-\text{CH}_2\text{OH}$  or  $-\text{CH}_2\text{OCH}_3$ ,

$\text{U}^2$  is radical  $\text{L}^1$ , whereby  $\text{L}^1$  and  $\text{U}^2$ , independently of one another, can be the same or different,

$\text{A}^1$  is a hydrogen atom, a straight-chain or branched  $\text{C}_1\text{-C}_{30}$  alkyl group, which optionally is interrupted by 1-15 oxygen atoms, and/or optionally is substituted with 1-10 hydroxy groups, 1-2  $\text{COOH}$  groups, a phenyl group, a benzyl group and/or 1-5  $-\text{OR}^9$  groups, with  $\text{R}^9$  having the meaning of a hydrogen atom or a  $\text{C}_1\text{-C}_7$  alkyl radical, or  $-\text{L}^1\text{-R}^{\text{F}}$ ,

  $\text{L}^1$  is a straight-chain or branched  $\text{C}_1\text{-C}_{30}$  alkylene group, which optionally is interrupted by 1-10 oxygen atoms, 1-5  $-\text{NH-CO}$  groups, 1-5  $-\text{CO-NH}$  groups, by a phenylene group optionally substituted by a  $\text{COOH-}$  group, 1-3 sulfur atoms, 1-2  $-\text{N}(\text{B}^1)\text{-SO}_2$  groups and/or 1-2  $-\text{SO}_2\text{-N}(\text{B}^1)\text{-}$ groups with  $\text{B}^1$  in the meaning of  $\text{A}^1$ , an  $\text{NHCO}$  group, a  $\text{CONH}$  group, an  $\text{N}(\text{B}^1)\text{-SO}_2$  group or an  $-\text{SO}_2\text{-N}(\text{B}^1)$  group and/or optionally is substituted with radical  $\text{R}^{\text{F}}$ , and

$\text{R}^{\text{F}}$  is a straight-chain or branched perfluorinated alkyl radical of formula  $\text{C}_n\text{F}_{2n}\text{E}$ , whereby  $n$  is number 4-30, and

$\text{E}$  is a terminal fluorine atom, chlorine atom, bromine atom, iodine atom or a hydrogen atom,

and optionally present acid groups optionally can be present as salts of organic and/or inorganic bases or amino acids or amino acid amides.

72. (NEW) A method according to claim 71, wherein the compound of formula Ib, is a compound in which  $\text{R}^2$ ,  $\text{R}^3$  and  $\text{R}^9$ , independently of one another, mean hydrogen or a  $\text{C}_1\text{-C}_4$  alkyl group.

73. (NEW) A method according to claim 71, wherein the compound of formula Ib, is a compound in which  $\text{A}^1$  is hydrogen, a  $\text{C}_1\text{-C}_5$  alkyl radical, or the radicals

$C_2H_4-O-CH_3$ ,  $C_3H_6-O-CH_3$ ,  
 $C_2H_4-O-(C_2H_4-O)_t-C_2H_4-OH$ ,  
 $C_2H_4-O-(C_2H_4-O)_t-C_2H_4-OCH_3$ ,  $C_2H_4OH$ ,  
 $C_3H_6OH$ ,  $C_4H_8OH$ ,  $C_5H_{10}OH$ ,  $C_6H_{12}OH$ ,  $C_7H_{14}OH$ ,  
 $CH(OH)CH_2OH$ ,  
 $CH(OH)CH(OH)CH_2OH$ ,  $CH_2[CH(OH)]_{u^1}CH_2OH$ ,  
 $CH[CH_2(OH)]CH(OH)CH_2OH$ ,  
 $C_2H_4CH(OH)CH_2OH$ ,  
 $(CH_2)_sCOOH$ ,  
 $C_2H_4-O-(C_2H_4-O)_t-CH_2COOH$  , or  
 $C_2H_4-O-(C_2H_4-O)_t-C_2H_4-C_nF_{2n}E$

whereby

s is integers 1 to 15,

t is integers 0 to 13,

$u^1$  is integers 1 to 10,

n is integers 4 to 20, and

E is hydrogen, fluorine, chlorine, bromine or iodine atoms, and optionally, their

branched isomers.

74. (NEW) A method according to claim 71, wherein the compound of formula Ib, is a compound in which  $A^1$  is hydrogen,  $C_1$ - $C_{10}$  alkyl,

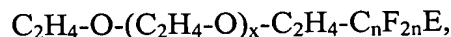
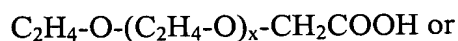
$C_2H_4-O-CH_3$ ,  $C_3H_6-O-CH_3$ ,

$C_2H_4-O-(C_2H_4-O)_x-C_2H_4-OH$ ,  $C_2H_4-O-(C_2H_4-O)_x-C_2H_4-OCH_3$ ,

$C_2H_4OH$ ,  $C_3H_6OH$ ,

$CH_2[CH(OH)]_yCH_2OH$ ,

$CH[CH_2(OH)]CH(OH)CH_2OH$ ,



whereby

x is integers 0 to 5,

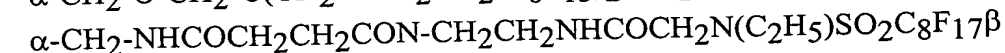
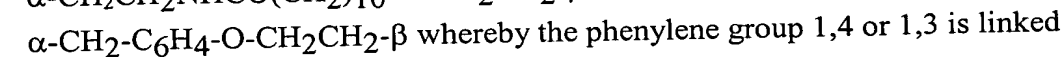
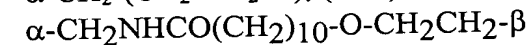
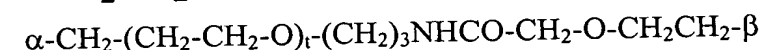
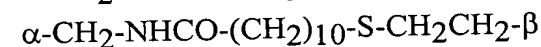
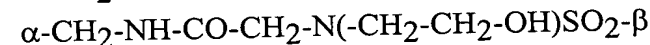
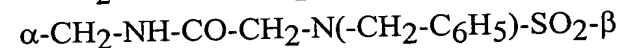
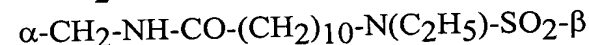
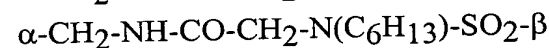
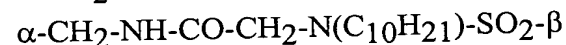
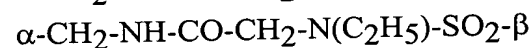
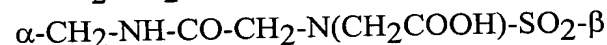
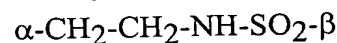
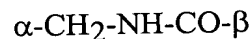
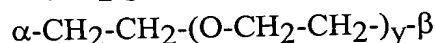
y is integers 1 to 6,

w is integers 1 to 10,

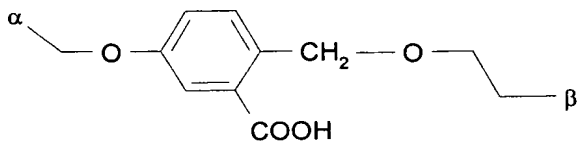
n is integers 4 to 15, and

E is a fluorine atom, and, optionally, their branched isomers.

75. (NEW) A method according to claim 71, wherein the compound of formula Ib, is a compound in which L<sup>1</sup> is



$\alpha\text{-CH}_2\text{-CH}_2\text{NHCOCH}_2\text{N}(\text{C}_2\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-O-CH}_2\text{-CH(OC}_{10}\text{H}_{21})\text{-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-(CH}_2\text{NHCO)}_4\text{-CH}_2\text{O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-(CH}_2\text{NHCO)}_3\text{-CH}_2\text{O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-OCH}_2\text{C(CH}_2\text{OH)}_2\text{-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$



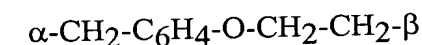
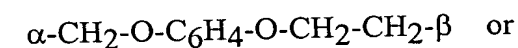
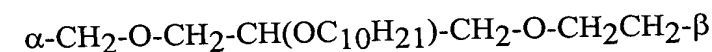
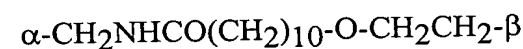
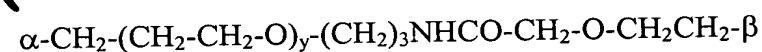
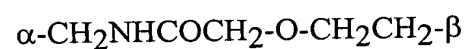
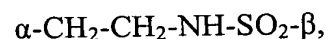
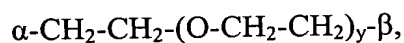
$\alpha\text{-CH}_2\text{NHCOCH}_2\text{N}(\text{C}_6\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-NHCO-CH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-NHCO-CH}_2\text{-O-CH}_2\text{CH}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(CH}_2\text{COOH)}\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_2\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_{10}\text{H}_{21})\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(C}_6\text{H}_{13})\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-(CH}_2\text{)}_{10}\text{-N(C}_2\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-C}_6\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-N(-CH}_2\text{-CH}_2\text{-OH)}\text{SO}_2\text{-}\beta$   
 $\alpha\text{-NH-CO-CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-O-C}_6\text{H}_4\text{-O-CH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-CH}_2\text{-C}_6\text{H}_4\text{-O-CH}_2\text{-CH}_2\text{-}\beta$   
 $\alpha\text{-N(C}_2\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_6\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_{10}\text{H}_{21})\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_6\text{H}_{13})\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-N(C}_2\text{H}_4\text{OH)}\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-N(CH}_2\text{COOH)}\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-N(CH}_2\text{C}_6\text{H}_5)\text{-SO}_2\text{-}\beta$   
 $\alpha\text{-N-[CH(CH}_2\text{OH)}_2]\text{-SO}_2\text{-}\beta$  or  
 $\alpha\text{-N-[CH(CH}_2\text{OH)CH(OH)(CH}_2\text{OH)]}\text{-SO}_2\text{-}\beta$

whereby

s is integers 1 to 15 and

y is integers 1 to 6.

76. (NEW) A method according to claim 71, wherein the compound of formula Ib, is a compound in which L<sup>1</sup> is



whereby

y is an interger from 1 to 6.

77. (NEW) A method according to claim 71, wherein the compound of formula Ib, is a compound in which R<sup>F</sup> is a straight-chain or branched perfluorinated alkyl radical of formula C<sub>n</sub>F<sub>2n</sub>E, whereby n is a number from 4 to 15 and E stands for a terminal fluorine atom.

78. (NEW) A method according to claim 71, wherein the compound of formula Ib



is a :

1,4,7-Tris(carboxylatomethyl)-10-(3-aza-4-oxo-hexan-5-yl)-acid-(2,3-dihydroxypropyl)-N-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl)-amide]-1,4,7,10-tetraazacyclododecane, gadolinium complex,

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)-acid-N-(3,6,9,12,15-pentaoxa)-hexadecyl)-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl]-amide]-1,4,7,10-tetraazacyclododecane, gadolinium complex,

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)-acid-N-5-hydroxy-3-oxa-pentyl)-N-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl]-amide]-1,4,7,10-tetraazacyclododecane, gadolinium complex,

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)-acid-[N-3,6,9,15-tetraoxa-12-aza-15-oxo-C<sub>17</sub>-C<sub>26</sub>-hepta-decafluor)hexacosyl]-amide]-1,4,7,10-tetraazacyclododecane, gadolinium complex, or

1,4,7-Tris(carboxylatomethyl)-10-[(3-aza-4-oxo-hexan-5-yl)-acid-N-(2-methoxyethyl)-N-(1H,1H,2H,2H,4H,4H,5H,5H-3-oxa)-perfluorotridecyl]-amide]-1,4,7,10-tetraazacyclododecane, gadolinium complex.

79. (NEW) A method according to claim 57, wherein the perfluoroalkyl-containing metal complex is in a galenical formulation that contains a paramagnetic, perfluoroalkyl-containing metal complex of formula I, and a diamagnetic perfluoroalkyl-containing substance, optionally dissolved in an aqueous solvent, wherein the diamagnetic perfluoroalkyl-containing substance is a compound of formula XX



in which R<sup>F</sup> is a straight-chain or branched perfluoroalkyl radical with 4 to 30 carbon atoms, L<sup>2</sup> is a linker and B<sup>2</sup> is a hydrophilic group.

80. (NEW) A method according to claim 79, wherein linker  $L^2$  is a direct bond, an  $-SO_2$  group, or a straight-chain or branched carbon chain with 1 to 20 carbon atoms, which can be substituted with one or more  $-OH$ ,  $-COO$ ,  $-SO_3$  groups and/or optionally contains one or more  $-O-$ ,  $-S-$ ,  $-CO-$ ,  $-CONH-$ ,  $-NHCO-$ ,  $-CONR^9$ ,  $-NR^9CO-$ ,  $-SO_2-$ ,  $-PO_4-$ ,  $-NH-$  or  $-NR^9$  groups, an aryl ring or a piperazine, whereby  $R^9$  is a  $C_1$  to  $C_{20}$  alkyl radical, which in turn can contain one or more O atoms, and/or can be substituted with  $-COO-$  or  $SO_3$  groups.


81. (NEW) A method according to claim 79, wherein hydrophilic group  $B^2$  is a mono- or disaccharide, with one or more adjacent  $-COO^-$  or  $-SO_3$  groups, a dicarboxylic acid, an isophthalic acid, a picolinic acid, a benzenesulfonic acid, a tetrahydropyrandicarboxylic acid, a 2,6-pyridinedicarboxylic acid, a quaternary ammonium ion, an aminopolycarboxylic acid, an aminodipolyethylene glycolsulfonic acid, an aminopolyethylene glycol group, an  $SO_2-(CH_2)_2-OH$  group, a polyhydroxyalkyl chain with at least two hydroxyl groups or one or more polyethylene glycol chains with at least two glycol units, whereby the polyethylene glycol chains are terminated by an  $-OH$  or  $-OCH_3$  group.

82. (new) A method according to claim 55, wherein the metal complex has a hydrodynamic micelle diameter of  $> 4$  nm.

83 (new) A method according to claim 56, wherein the metal complex has a proton relaxivity in plasma of  $> 15$  l/mmol's.

84. (new) A method according to claim 61, wherein the perfluoroalkyl-containing metal complex is in a galenical formulation that contains a paramagnetic, perfluoroalkyl-containing metal complex of formula Ia and diamagnetic perfluoroalkyl-containing substance, optionally dissolved in an aqueous solvent.

85. (new) A method according to claim 71, wherein the perfluoroalkyl-containing metal complex is in a galenical formulations that contains a paramagnetic, perfluoroalkyl-containing metal complex of formula Ib, and a diamagnetic perfluoroalkyl-containing substance, optionally dissolved in an aqueous solvent.

 86. (new) A method according to claim 51, wherein plaque is visualized.

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